EXTRACTION OF ESSENTIAL OILS FROM PATCHOULI LEAVES USING ULTRASONIC-ASSISTED SOLVENT EXTRACTION METHOD

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"Saya akui bahawa saya telah membaca karya ini dan pada pandangan saya karya ini adalah memadai dari segi skop dan kualiti untuk tujuan penganugerahan ijazah Sarjana Muda Kejuruteraan Kimia.".

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I declare that this thesis entitled "*Extraction of Essential Oils from Patchouli Leaves Using Ultrasonic-Assisted Solvent Extraction Method*" is the result of my own research except as cited in the references. The thesis has not been accepted for any degree and is not concurrently submitted in candidature of any other degree.

Signature : .....

Name of Candidate: MUHD ZAHIRUDDIN BIN SHUKOR

Date : 14 MAY 2008

Special Dedication of This Grateful Feeling to My…

Beloved father and mother;
Mr. Shukor Mohd Nordin and Mrs. Zaitun Ibrahim

Loving brothers and sisters;
Zuhairi, Fadhli, Aiman, Faiz, Fahmi, Syazwani

Supportive families;
Uncles and Aunties

For Their Love, Support and Best Wishes.

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ABSTRACT

Essential oil is an aromatic liquid that is extracted from various parts of the plants. It contains the true essence of the plant and has many therapeutic benefits. Patchouli essential oil from the extraction of dried Patchouli (*Pogostemon Cablin*) leaves is the important ingredient in many fragrance products like perfumes and also use widely in medical field. This experiment use ultrasonication-assisted solvent extraction method that comprises two set of experiments in order to investigate the effect of ultrasonic and type of solvent on extraction process. Ethanol, hexane and acetone are the solvents used for the first experiment. The best solvent among three is chosen to be used in second experiment. In the second experiment, ultrasound is used in order to investigate its effect compared to the experiment without using ultrasonic. The qualitative and quantitative analysis has been done in order to show the objectives were achieved. Qualitative analysis involved the chromatogram analysis from GCMS while quantitative analysis is based on the percent yield. From qualitative analysis, ethanol gives the highest peak area (27.92%) than hexane (20.01%) and acetone (20.42%). In addition, average peak area for ultrasonic method (50.18%) is better than without using ultrasonic (42.40%). Meanwhile, for qualitative analysis, ethanol can extract highest yield (2.87%) compared to hexane (2.53%) and acetone (2.00%). Then, by using ultrasonic, it gives higher average yield (2.27%) than without using ultrasonic (1.67%). Therefore, from these analyses, the best solvent used for solvent extraction is ethanol because it produced highest quality and most yields of patchouli oil. This experiment also has the better result when it involves the ultrasonication method.

ABSTRAK

Pati minyak adalah cecair aroma yang boleh diekstrak daripada banyak bahagian tumbuhan. Ia mengandungi pati asli tumbuhan dan mempunyai banyak kelebihan untuk terapi. Minyak patchouli yang terhasil daripada pengekstrakan daun (*Pogostemon cablin)* kering merupakan bahan ramuan dalam banyak produk wangian seperti minyak wangi dan juga banyak digunakan dalam bidang perubatan. Eksperimen ini menggunakan kaedah penggunaan pelarut dengan bantuan ultrasonik yang melibatkan dua set eksperimen dalam usaha untuk mengkaji kesan ultrasonik dan pelarut dalam proses pengekstrakan. Etanol, heksane dan aseton adalah tiga pelarut yang digunakan dalam eksperimen ini. Pelarut yang terbaik daripada tiga pelarut tersebut akan dipilih untuk digunakan dalam eksperimen yang kedua. Dalam eksperimen yang kedua, ultrabunyi digunakan untuk mengkaji kesannya jika dibandingkan dengan eksperimen yang tidak menggunakan ultrasonik. Analisis kualitatif dan kuantitatif telah dibuat untuk menunjukkan objektif tercapai. Analisis kualitatif melibatkan analisis kromatogram daripada GCMS manakala analisis kuantitatif pula berdasarkan peratus hasil. Daripada analsis kualitatif, etanol memberikan luas puncak yang paling besar (27.92%) berbanding heksane (20.01%) dan aseton (20.42%). Selain itu, purata luas puncak kaedah penggunaan ultrabunyi (52.18%) lebih baik daripada tidak menggunakan ultrabunyi (42.40%). Manakala daripada analisis kuantitatif, etanol boleh mengektrak hasil yang paling banyak (2.87%) berbanding heksane (2.53%) dan aseton (2.00%). Kemudian, penggunaan ultrabunyi memberikan lebih purata hasil (2.27%) berbanding tidak menggunakan ultrabunyi (1.67%). Maka, daripada analisis ini etanol adalah pelarut terbaik kerana ia menghasilkan hasil minyak patchouli yang paling berkualiti dan paling banyak. Daripada eksperimen ini juga, hasil minyak adalah lebih baik sekiranya melibatkan penggunaan ultrasonik.

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LIST OF SYMBOLS

%	- Yield percentage
°C	- Degree Celsius
µ	- Micro

LIST OF ABBREVIATIONS

g	- Gram
ml	- Mililiter
GCMS	- Gas Chromatography Mass Spectrometer
Min	-Minute

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CHAPTER 1

INTRODUCTION

1.1 Background

Essential oil is a concentrated liquid that contains various elements such as aromatic compounds, organic constituents, including hormones, vitamin and other natural elements. These compounds are extracted from various parts of a plant and are highly volatile. In the plant, essential oils are produced inside the protoplasm of the cells and stored as micro droplets in the glands of the plant. The oil needs to diffuse through the wall of the glands and spread over the surface of the plant before evaporating and filling the air with perfume.

The oils are rich in energy and chemically active. As such, essential oils are used for many different reasons and in different ways. Commercially, essential oils are used extensively in three main industries which are food, pharmaceutical and fragrance industries. Modern scientific research leads to production synthetic essential that creates the fragrances. However, they are dissimilar from natural fragrance oils or perfume as essential oils are derived from the true plants, as such it may also not contain the therapeutic benefit same like the natural essential oil does.

Patchouli essential oil is obtained from the leaves of *Pogostemon cablin* (Patchouli), a plant from the *Lamiaceae* family, and is widely acknowledged for its characteristic pleasant and long lasting woody, earthy, camphoraceous odor. It is one of the important natural essential oils used to give a base and lasting character to a fragrance in perfumery industry. Patchouli oil is an important ingredient in many fragrance products like perfumes, as well as in soaps and cosmetic products (Bauer *et al.*, 1997) as well as for aromatherapy, spiritual use and medicinal field.

The composition of the patchouli oil is complex but distinct because it consists largely of sesquiterpenes (Buré *et al.*, 1719; Dung *et al.*, 1989; Lawrence, 1990). Specifically, the main constituent of patchouli oil is patchoulol. At present, patchouli plants are the only commercial source of patchoulol and cost-effective synthetic routes for enantiomeric pure patchoulol have yet to be developed (Näf *et al.*, 1981; Srikrishna and Satyanarayana, 2005).

From the researches that have been done before, there are many methods to extract essential oil including conventional and modern techniques but each of them has its own advantages and disadvantages. The suitable method depends on the plant that we want to extract. So, the selection of the best method is crucial to ensure the best quality of essential oil obtained. Most oils are extracted using steam distillation, which is one of the conventional methods.

In this experiment, we study on two methodologies which are the solvent extraction and ultrasonication. These two methods are independent and have the different scope of experiment. Solvent extraction uses the solvent as the medium extractor while ultrasonication use wave to ease the extraction of essential oils from patchouli leaves. Then, the yield from both experiments will be analyzed to obtain the compound in the patchouli oil. Finally, qualitative and quantitative analysis will be done to show the objectives are achieved.

1.2 Problem Statement

In this study, the patchouli leaf is being used as the raw material. Currently, the demand of patchouli oil is very high almost every year (2000 tonnes per annum) because it has wide range of usages. Therefore, it is a big potential to make the profit from this patchouli oil extraction as the growth of market demand. Even many aromatics chemicals have been produced, many people still prefer to the true botanical aroma.

Patchouli is commonly extracted for its essential oils using methanol or ethanol as solvent. This time we are using other solvents which are ethanol, hexane and acetone to make comparison and choose the best solvent in extracting patchouli essential oils. Conventional steam distillation method use a large amount of heat in the process which can cause the thermal degradation of many compounds contained in the patchouli leaf. Compared to steam distillation method, solvent extraction method is more suitable to be used on delicate plants because it uses very little heat that makes it able to produce essential oils in higher amounts and at lower cost. Other than that, it is important to improve the existing products of fragrance and also try to encourage the development of local technologies to take advantage of market opportunities.

Meanwhile, ultrasonication is the new method in essential oil extraction. The wave used will penetrate the cell walls and facilitates the transfer from the cell into the solvent. Therefore, the extraction becomes easier. So, this research is important in comparing the effect of using solvent extraction and ultrasonic extraction method to the extraction process of patchouli oil.

1.3 Objective

The aim of this project is to extract essential oils from patchouli leaves by using ultrasonic-assisted solvent extraction method.

1.4 Scope

This research is based on experimental studies of solvent extraction and ultrasonication. In order to achieve the objectives mentioned above, three scopes have been identified:

i. To investigate the effect of solvents on extraction.
ii. To investigate the effect of ultrasonic on extraction.
iii. To analyze the product compounds from the extraction process.

CHAPTER 2

LITERATURE REVIEW

2.1 Essential Oils

2.1.1 Introduction

Essential oil is known as volatile or ethereal oils, or simply as the oil of the plant material from which they were extracted. The term essential shows that the oil carries distinctive scent (essence) of the plant. Essential oils do not as a group needs to have any specific chemical properties in common, beyond conveying characteristic fragrances. They are 75 to 100 times more concentrated than the oils in dried herbs. The use of volatile plant oil, including essential oils, for psychological and physical well being has dated back thousands of years. The ancient Chinese are generally acknowledged as the founders of aromatherapy from essential oils and it was used by the ancient Egyptians and ancient Greeks as medicinal perfumes.

Essential oil can be generally distilled from the leaves, stems, flowers, bark, roots or other elements from various parts of plants. They are not true oils in the manner of lubricant vegetable oils, but highly fluid and exceptionally volatile. However, most of essential oils are clear and contain the true essence of the plant it was derived from. Experts recognize an essential oil by its aroma and check its composition by a process called Gas Liquid Chromatography. Colour can also be an indicator; eucalyptus is colourless, chamomile varies from white to blue and others,

like basil and sandalwood (both light greenish-yellow), are in pastel shades. Yet others are richly pigmented, like jasmine, a deep reddish-brown, patchouli, brown, and rose, orange-red.

There are only about 700 plants which considered aromatic among all types of plants in the world that can be used for the production of essential oils and hundreds of other essential oils available for use, many with known antibacterial properties. Table 2.1 shows the parts of various plants that can be extracted.

Table 2.1: Parts of various plants for extraction

Berries • Allspice • Juniperkejut Seeds • Almond • Anise • Celery • Cumin • Nutmeg oil Bark • Cassia • Cinnamon • Sassafras Wood • Camphor • Cedar • Rosewood • Sandalwood Rhizome • Ginger	Leaves • Basil • Bay leaf • Cinnamon • Common sage • Eucalyptus • Lemon grass • Melalueca • Oregano • Patchouli • Peppermint • Pine • Rosemary • Spearmint • Tea tree • Thyme • Wintergreen Resin • Frankincense • Myrrh	Flower • Chamomile • Clary sage • Clove • Geranium • Hyssop • Jasmine • Lavender • Manuka • Marjoram • Orange • Rose • Ylang-ylang Peel • Bergamot • Grapefruit • Lemon • Lime • Orange • Tangerine Root • Valeria

2.1.2 Essential Oils Constituents

Essential oils contain numerous constituents that contribute to the characteristic odour and medicinal effects. The major chemical components that account for the pleasant aromatic odours are primarily terpenenes, monoterpenes and linalool (Williams, 1997). The presence and quantity of the various components varies between oils and determines the individuality of the oil (Tisserand and Balacs, 1996).

Essential oils represent a small fraction of a plant's composition but confer the characteristic for which aromatic plants are used in the pharmaceutical, food and fragrance industries. Essential oils have a complex composition, containing from a few dozen to several hundred constituents, especially hydrocarbons (terpenes and sesquiterpenes) and oxygenated compounds (alcohols, aldehydes, ketones, acids, phenols, oxides, lactones, acetyls, ethers and esters). Both hydrocarbons and oxygenated compounds are responsible for the characteristic odors and favors. The proportion of individual compounds in the oil composition is different from trace levels to over 90%. The aroma's oil is the result of the combination of the aromas of all components. Trace components are important, since they give the oil a characteristic and natural odor. Thus, it is important that the natural proportion of the components is maintained during extraction of the essential oils from plants by any procedure (Anitescu *et al.*, 1997).

2.1.3 Uses of Essential Oils

Various essential oils have been used medicinally at different periods in history. Medical applications proposed by those who sell medicinal oils vary from skin treatments to remedies for cancer, and are often based on historical use of these oils for these purposes. Interest in essential oils has revived in recent decades, with the popularity of aromatherapy, a branch of alternative medicine which claims that the specific aromas carried by essential oils have curative effects. Oils are volatilized or diluted in carrier oil and used in massage, or burned as incense, for example.

Furthermore, these aromatic characteristics of essential oils may provide various functions for the plants itself including; attracting or repelling insects (odors of the flowers); while in plant metabolism, a few essential oils might involved in this process; Leaf oils, wood oils, and root oils may serve to protect against plant parasites or depredations by animals as well as anti-bacterial agent which is utilizing the hormone in the oil.

2.1.4 Essential Oil versus Synthetics Oils

Nowadays, essential oils are subjected to more scientific investigation and it was discovered that some of them could be synthesized from other materials. Essential oils are not the same as fragrance oils or perfume where essential oils are derived from the true plants.

Synthetic essential oils are produced by blending aromatic chemicals mostly derived from coal tar which can duplicate the smell of the pure essential oils while the essential oil have the complex chemical components which created in nature that can determine its true aromatic benefits. Synthetic essential oils are unnaturally created fragrances since it contains artificial substances, and it may also do not offer the therapeutic benefit that essential oils does. This is how we can easily compare the differences between synthetic and pure essentials oils. Although synthetic essential oils are not suitable for aromatherapy, they still can be used as the scent to crafts, potpourri, soap and perfume at a fraction of the cost. The reason of these synthetic products is mainly to reduce the cost of production. As it is always quicker and cheaper to produce the laboratory versions than natural plant extracts, true essential oils began to fall from favor.

There is no synthetic substitute for patchouli oil until today, which increases its value and demand in the perfumery market. Currently, the consumption of patchouli oil in the world is about 2000 tonnes per annum (Amir H. Barati *et al.*, 2007).

2.1.5 Hazardous of Essential Oils

Although essential oils are known for their antimicrobial properties, medical teams rarely use them. This is primarily due to lack of scientific evidence of their efficacy, toxicity issues and the availability of conventional therapy. Most readily available essential oils are safe if used in small doses, and side effects are generally rare. Possible side effects include rashes, itching, and irritation on the skin. Allergic reactions include watery eyes, sneezing, and inflammation. Some essential oils may cause nausea, dizziness, or gastrointestinal discomfort when used in excess or by those with allergic reactions. Some essential oils, particularly those derived from citrus fruit plants, can cause increased sensitivity to sunlight and increased risk of sunburn. In addition, some essential oils have not been thoroughly tested and may be toxic. Therefore, any essential oils that have not been tested or lack adequate information should be avoided.

2.2 Patchouli

2.2.1 Patchouli Plant

Patchouli is an aromatic herb plant. Patchouli is native to The Philippines and grows wild and also cultivated in Malaysia, Indonesia, Singapore, China and India. This fragrant herb is a bush with furry leaves and purplish white flowers. It can grow up to three and half feet tall and it has large fragrant leaves. True patchouli has hairy stems, flowers only reluctantly, and is usually propagated by cuttings. Figure 2.1 and Table 2.2 show the images and the scientific classification of patchouli plant.

Figure 2.1: Patchouli *(Pogostemon cablin)*

Table 2.2: Scientific classification of patchouli

Kingdom	Plantae
Division	Magnoliophyta
Class	Magnoliopsida
Order	Lamiales
Family	Lamiaceae
Genus	*Pogostemon*
Species	*P. cablin*

Patchouli is a plant which has good economic prospect. Patchouli leaves are the economic part that contains the oil gland to be extracted out. The leaves of the patchouli bush are first dried in the sun, and then distilled to produce thick oil, amber to dark orange in colour (Fang Chen *et al.*, 2007). The yield of oil from the dried leaves is about 3%. Natural fragrances like sandalwood, rose, jasmine, vetiver, agarwood and patchouli are complex mixtures of organic molecules, which cannot be reproduced in the laboratory. Thus, patchouli enjoys an additional importance as aromatic oil. Patchouli alcohol will have long-lasting fragrant aroma when blended with other aroma chemicals.

The world production is estimated to be more than 500 tonnes/year. A small number of companies have specialized in the manufacture of refined qualities of patchouli oil for the perfume industry, where it finds extensive use in modern high-

class perfumes. There are no synthetic equivalents of the patchouli scent. The shade dry leaf upon steam distillation yields the patchouli oil of commerce, which is used in perfumery, cosmetics, processed food and is imported into India every year in large quantities. The essential oil is one of the best fixatives for heavy perfumes, which imparts strength, strong character, alluring notes and lasting patchouli qualities.

2.2.2 Patchouli Essential Oil

Patchouli oil is one of the important natural essential oils used to give a base and lasting character to a fragrance in perfumery industry. The dry leaves of patchouli when extracted using steam distillation yield an essential oil called the oil of patchouli. Indonesia is the major producer of patchouli oil in the world with an estimated 550 tons per year, which is more than 80% of the total (Robbins, 1983; Tao, 1983).

The essential oil of patchouli is extracted from the leaves. The leaves need to be shade dried and partially fermented before distilling. Fresh patchouli essential oil has a sharp, green fragrance, and needs to age to develop the deeper, earthier aroma of good patchouli oil. Patchouli essential oil should always be aged and will continue to improve the longer it sits. The color of the oil will deepen from a light yellowish, pale red to deep, dark amber upon aging, and the oil will become more and more viscous. It is non-toxic, non-irritant and non-sensitizing, but the smell of patchouli oil may be a little persistent for some people and large doses may cause loss of appetite in some individuals. There is no synthetic substitute for patchouli oil until today, which increases its value and demand in the perfumery market.

2.2.2.1 Patchouli Essential Oil Constituents

The chemical components of Patchouli oil are b-patchoulene, a-guaiene, caryophyllene, a-patchoulene, seychellene, a-bulnesene, norpatchoulenol, Patchouli alcohol and pogostol. The constituents of the oil include: Patchoulol (25-35%), Alpha-Bulnesene (12-20%), Alpha-Guaiene + Seychellene (15-25%), and Alpha-Patchoulene (5-9%) (Srikrishna and Satrayanarayana, 2005). However, it is maintained that norpatchoulenol, present in only 0.3-0.4 %, is playing a principal part in the overall odor picture. Natural patchouli oil contains numerous other compounds, including a wide variety of hydrocarbons, epoxides, alcohols and sesquiterpene ketones many of which are also odoriferous compounds. Figure 2.2 shows the structures of nine identified compounds in patchouli oil.

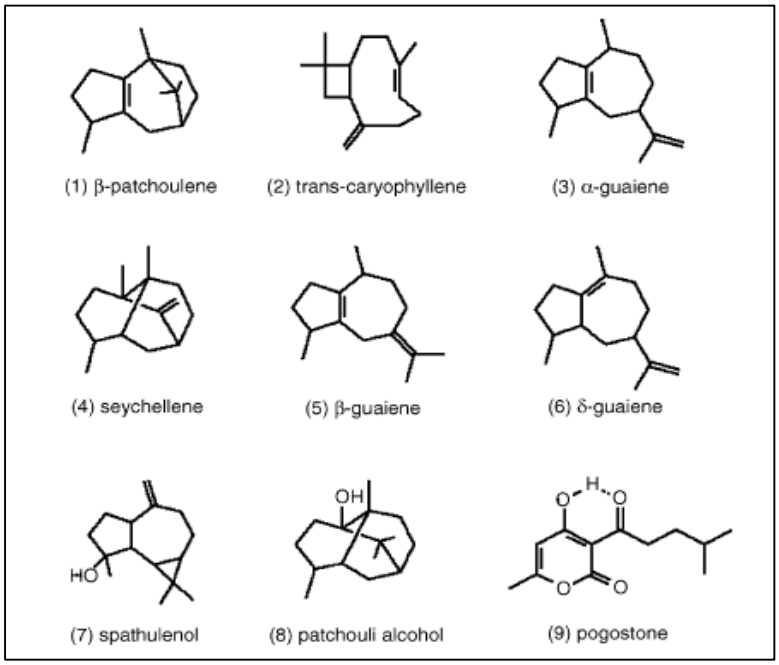

Figure 2.2: The structures of nine identified compounds in *Pogostemon cablin*.